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U1S 1462 1626 B5A C3C

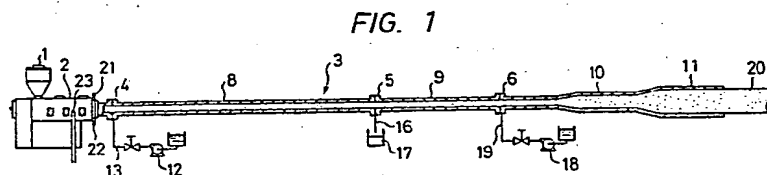
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(57) Thermoplastic resin containing a crosslinking agent and a foaming agent is extruded through a long-land die 3 which is provided with an applicator 4 for applying lubricant onto the inner surface of the long-land. A lubricant extractor 5 for removing the lubricant and, optionally a further lubricant applicator 6, are provided downstream of the applicator 4. The crosslinking agent is decomposed in the front part 8 of the die 3 to lower the flow rate of the resin and the foaming agent is decomposed or heated to its boiling point in the rear parts 9, 10 of the die 3 to provide an expandable resin. The crosslinked, expandable resin is then released into an atmospheric or controlled pressure zone 11 to produce a uniform, finely expanded foam article.



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FIG. 1

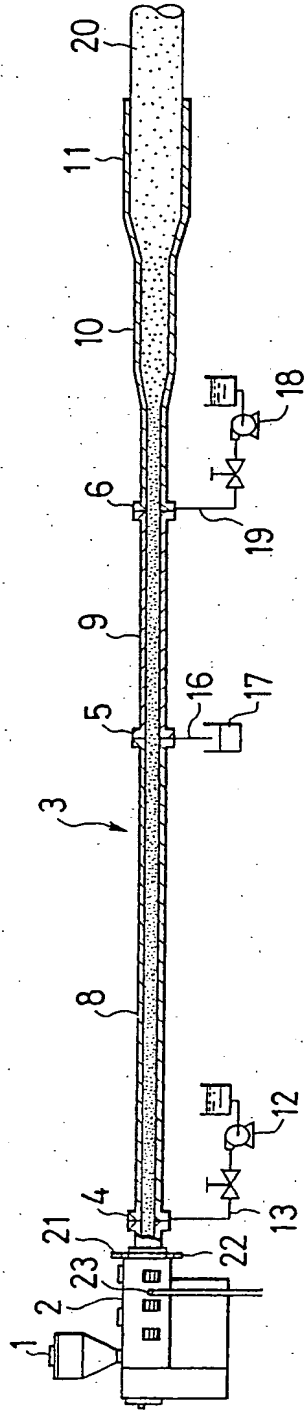


FIG. 2

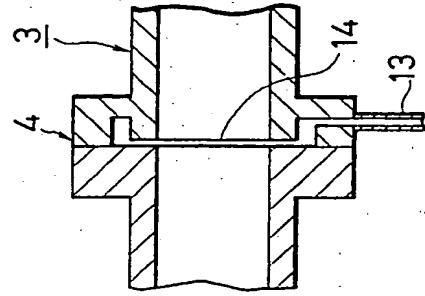


FIG. 3

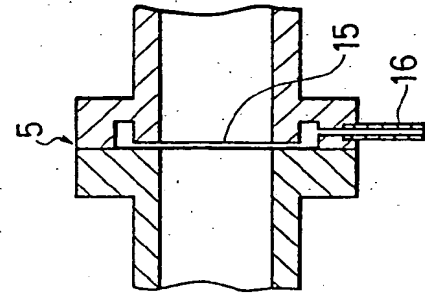


FIG. 4

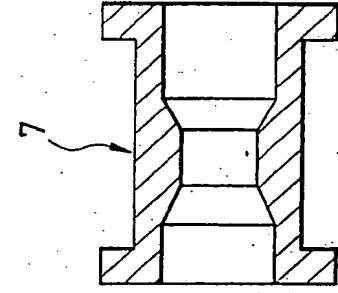
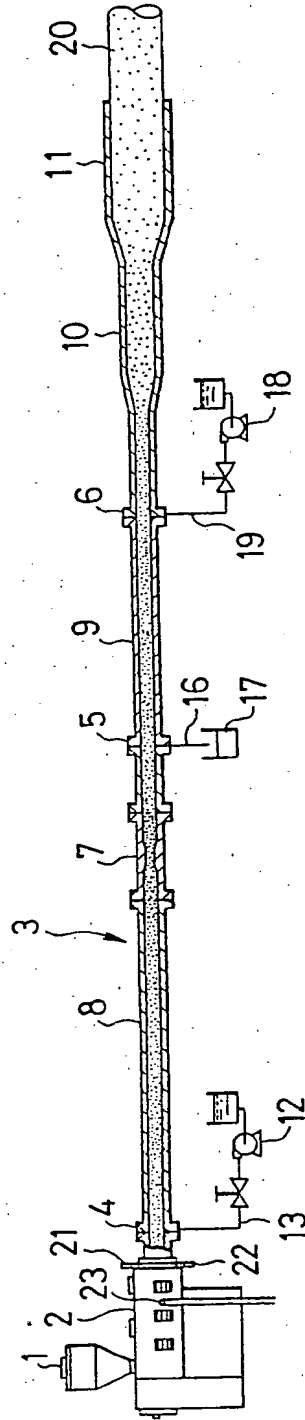


FIG. 5



## SPECIFICATION

## Method of producing crosslinked foam articles

5 This invention relates to a method of producing uniformly expanded crosslinked foam articles by extrusion-forming.

Various methods of producing foamed thermoplastic articles by continuous extrusion are known; and, as the foaming agent, both physical agents which volatilize by heating and chemical agents which decompose and generate gas by heating have been used. However, it has been difficult to obtain uniform, fine-celled, highly expanded foam from thermoplastic resins because such resins decrease sharply in viscosity with heating and melting.

In order to avoid this defect, a crosslinking foaming process wherein a thermoplastic resin is crosslinked to adjust its melt viscosity to a suitable value for foaming has been proposed. For example, a thermoplastic resin comprising a chemical foaming agent and a crosslinking agent which has a lower decomposition temperature than the chemical foaming agent is formed into a desired shape. The shaped article is heated at atmospheric pressure or under controlled pressure, firstly, to cause crosslinking by decomposition of the crosslinking agent and, secondly, to cause expansion of the resin by decomposition of the chemical foaming agent.

At atmospheric pressure, rapid foaming causes foaming of only the surface portion of the article. The foamed surface portion acts as an insulation layer and prevents thermal conduction to an inner portion of the resin and the chemical foaming agent cannot decompose smoothly. Even if a resin compound is heated slowly, decomposition of a chemical foaming agent in the innermost portion of an article thicker than 10 mm is difficult.

Accordingly, the upper limit of the thickness of such a foamed article is about 20 mm and a foamed article thicker than 20 mm is usually made by lamination.

On the other hand, for the production of crosslinked foamed articles under pressure there are known a batchwise method and a continuous method. In the batchwise method, as described in the Japanese Patent Publication No. 29381/70 (Yoshida et al), a thermoplastic resin compound comprising a crosslinking agent and a foaming agent is heated in a mold through one step or two steps to cause crosslinking and foaming of the resin. In this process, even though thick, uniform, fine-celled, foamed articles can be obtained, productivity remains low due to the use of a batchwise system.

In the continuous method, Japanese Patent Laid Open Application No. 1531/83 (Matsui et al) shows a method of foaming a polyolefin in which a polyolefin containing an organic peroxide and foaming agent is extruded into a long-land die, formed into a tubular shape, crosslinked and exposed to a lower pressure zone than in the long-land die to cause expansion. This process uses an oil lubricat-

ing method as described in U.S. Patent No. 3,928,525 (Matsui et al) in which a thermoplastic resin is crosslinked in a long-land die and extruded continuously therefrom. In this process, however, bubbles which are generated from the foaming agent cause thermal conduction to an inner portion of the resin to reduce and prevent uniform crosslinking.

Many kinds of foaming agents normally used in a crosslinking-foaming process may generate bubbles in a resin by volatilizing or decomposing partially during crosslinking to decrease the flow rate of the resin and, as a result, to decrease thermal conduction and retard the crosslinking reaction. If bubbles are generated in a resin before the flow rate of the resin becomes sufficiently low due to crosslinking, the dimensions of cells in the finally produced foamed article are extremely nonuniform.

The inventors proposed a method which overcomes the aforesaid problems and continuously produces crosslinked foamed articles (Japanese Patent Application No. 52548/81). The method comprises extruding a resin containing a foaming agent and a crosslinking agent into an oil lubricated long-land die, heating, crosslinking and decomposing the foaming agent, wherein a choking device is provided at the end and/or intermediate portions of the long-land die so that the resin in the long-land die is subjected to a back pressure. Homogeneous dissolution of the gas generated from the foaming agent into the resin is maintained and crosslinking is conducted while preventing separation and bubbling of the gas; as a result, release of the resin from the long-land die into a lower pressure zone results in a uniformly, finely foamed article being obtained. However, this method requires changes in the structure of the choking orifice depending upon the type of resin and foaming agent and the desired degree of foaming.

The inventors have now found a method which does not require the choking orifice and in which, even in the production of a thick article, uniform expansion can be achieved in the innermost portion.

According to the present invention a method of producing a cross-linked foam article comprises extruding a crosslinkable thermoplastic resin containing a foaming agent and a crosslinking agent into a long-land die wherein at an end portion and/or an intermediate portion of the long-land die a lubricant extracting device is provided; feeding a lubricant onto the inner surface of the long-land die; heating the resin to cause crosslinking and decrease of the melt flow rate of the resin by decomposition of the crosslinking agent; and thereafter expanding the resin by decomposition of the foaming agent. By providing a lubricant extraction device at an end or intermediate portion of the long-land die, the resin in the long-land die is subjected to a back pressure. Separation of gas generated from the foaming agent is thereby prevented and homogeneous dissolution of the gas into the resin occurs during the process of crosslinking.

Release of the resin from the long-land die into a lower pressure zone results in a uniformly, finely foamed article being obtained.

By the method of the present invention, thick and finely foamed articles of high to low expansion rates can be made with high productivity and at an unpredictably improved extruding speed.

In the drawings of the accompanying specification:

Figure 1 is a side view, partly in section, of an apparatus for carrying out the method of the invention,

Figure 2 is a sectional view of a lubricant applicator used in the method of the invention,

Figure 3 is a sectional view of a lubricant extracting device used in the method of the invention,

Figure 4 is a sectional view of a choking orifice used in the method of the invention, and

Figure 5 is a side view of an apparatus for carrying out another embodiment of the method of the invention.

Crosslinkable thermoplastic resins useful in the method of the present invention are not particularly limited and include high density and low density polyethylene, polystyrene, polypropylene, polyvinylchloride, polyamide and copolymers in which the aforesaid polymers are present in a predominant amount such as, for example, ethylene-propylene copolymer, ethylene-vinylacetate copolymer, ethylene-vinylchloride copolymer, vinylchloride-vinylidenechloride copolymer and mixtures of these polymers and copolymers. Elastomers such as EPR, EPDM, SBR, IIR and rubber can be used also. Furthermore, the thermoplastic resins can contain various additives such as paraffin, another thermoplastic resin, plasticizer, pigment, flame retardant agent, antistatic agent and stabilizers as will be appreciated by those skilled in the art.

Polyolefins and other crystalline thermoplastic resins have particular utility in the method of the invention and can be made into crosslinked foamed articles having uniform fine cells and low to high expansion rates. Moreover, even high density polyethylene and polypropylene which are not widely used in the prior art methods can be made into thick crosslinked foamed articles.

Crosslinking agents which can be used in this invention include any of those agents suitable for cross linking the thermoplastic resins and which have a higher decomposition temperature than the melt initiating temperature of the resin.

Typical crosslinking agents are, for example, organic peroxides such as ditertiarybutyl peroxide, tertiarybutylcumyl peroxide, dicumyl peroxide,  $\alpha$ - $\alpha$ -bis(tertiarybutyl peroxy diisopropyl)benzene, 2,5-dimethyl 2,5-di(tertiarybutyl peroxy)hexane, 2,5-dimethyl-2,5 (tertiarybutyl peroxy)hexane-3 or mixtures thereof and sulfur. If desired, a retarder or an accelerator for crosslinking can be added as an auxiliary agent.

The "decomposition temperature" of the cross linking agent means the temperature at which the half-life period of the agent becomes less than ten minutes. The quantity of crosslinking agent used should be determined according to various factors

such as molecular weight, molecular weight distribution, extent of branching of the resin used, degree of crosslinking desired and the decomposition temperature of the crosslinking agent. Usually the quantity of the crosslinking agent is 0.1-5 wt%, and, preferably, 0.2-2 wt% based on the weight of the crosslinkable thermoplastic resin composition.

If the amount of crosslinking agent used is less than 0.1 wt%, lowering of the melt flow rate of the resin, especially of a crystalline thermoplastic resin, by crosslinking is insufficient and, as a result, it is difficult to make a thick crosslinked foam article having a uniform cellular structure. On the other hand, when more than 5 wt% of a crosslinking agent is used, the melt flow rate is reduced excessively in the extruder whereby a uniform cellular structure of the crosslinked foamed article is difficult to obtain.

As the foaming agent in the method of this invention, virtually any volatile physical foaming agent or chemical foaming agent which generates gas such as nitrogen, carbon dioxide or ammonia by decomposition can be used. Volatile physical foaming agents include aliphatic hydrocarbons, for example, butane, pentane, hexane, heptane and halogenated aliphatic hydrocarbons such as methyl chloride, methylfluoride and tetrafluoro ethylene. As for chemical foaming agents, the decomposition temperature should be higher than that of the crosslinking agent used. For example, azodicarbonamide, metal salt of azodicarbonamide, dinitrosopentamethylene tetramine, azobis-isobutyronitrile, trihydrazinotriazine, 4,4-oxybis-benzenesulfonylhydrazide, 4,4-oxybis-benzen sulfonylsemicarbazide can be used. These foaming agents can be used as mixture of two or more than two kinds of agents and various kinds of auxiliary agents can be added as will be understood by those skilled in the art.

The method of the present invention can be better understood by referring to the accompanying drawings which are schematic representations of specific embodiments of apparatus useful in carrying out the method of the invention.

In Fig. 1, a crosslinkable thermoplastic resin containing a crosslinking agent and a foaming agent is fed into hopper 1 of extruder 2 and mixed and kneaded in the extruder at a temperature where there is no substantial decomposition of said crosslinking agent and foaming agent and then is extruded into long-land die 3. A thermometer, or thermocouple 21, and a pressure gauge 22, are provided at the outlet of the extruder 2 to measure the resin temperature and pressure. At a position adjacent to the entrance of the long-land die, lubricant applicator 4 is provided to supply lubricant to the inner surface of the long-land die 3.

An example of a lubricant applicator is shown in Fig. 2. Lubricant applicator 4 is provided with slit 14 in its inner surface and conduit pipe 13 which is connected to feeding pump 12 and lubricant reservoir. Lubricant can be fed and spread through slit 14 into the interface between the inner surface of the long-land die and the surface of the resin being extruded, forming a continuous film of lubricant.

As crosslinking of a resin by decomposition of a

crosslinking agent progresses, the flowability of the resin decreases and friction of the resin increases and finally it becomes very difficult to form the resin into a desired shape, and damage of an extruder may occur. The lubricant fed through applicator 4 eliminates this problem and makes it possible for the crosslinked resin to flow smoothly through the long-land die.

As the lubricants which can be employed in the present invention, chemically stable substances which do not decompose, boil, accelerate degradation of the thermoplastic resin and which are not soluble in the resin are preferred. For example, polysiloxanes such as polydimethyl siloxane and polydimethylsiloxane, polyhydric alcohols and their alkylesters and their alkylethers, and polyoxyalkylenes and their derivatives such as random, block or graft copolymers of two or more alkylene oxides can be used. Preferably, a water soluble surfactant such as a polyhydric alcohol is recommended because the lubricant can easily be removed from the surface of the foamed article by washing.

In the body of the long-land die, a heater (not shown) which heats the resin at a controlled temperature; preferably, an electrical band heater, is provided.

In front part 8 of long-land die 3, the crosslinking agent is decomposed at a temperature lower than the decomposition temperature of the foaming agent to cause the thermoplastic resin to crosslink and to thereby decrease the melt flow rate of the resin.

The degree of crosslinking of the resin is measured for example by dissolving the resin in boiling xylene. In the present invention, the gel ratio which is defined by the weight % of residue gel which remains undissolved after a 10 hour extraction by boiling xylene is more than 5% and, preferably 10-70%, whereby the viscosity of the resin is adjusted suitably and almost all of the gas generated from the foaming agent can be effectively utilized for expansion.

The length of the long-land die is determined by parameters such as resin temperature, foaming speed, heat conductivity of the resin and decomposing characteristics of the crosslinking agent. Usually the length of front part 8 is more than 50 cm, preferably more than 200 cm. With a long-land die shorter than 50 cm it is difficult to obtain a uniform cellular article. The optimum length of the die should be determined by taking into account the production rate and economical factors such as cost of installation or utilities.

A lubricant extracting device 5 having the structure as shown in, for example, Fig. 3 is provided at the position of the long-land die having a length sufficient to substantially complete the crosslinking of the resin, thereby removing the lubricant on the surface layer of the resin fed in the long-land die 3.

The resin from which the lubricant has been removed increases the surface resistance by decreasing the smoothness to the long-land die, thereby applying a back pressure into the long-land die 8, 9.

A part of the foaming agent foams by volatilization or decomposition even at the temperature in the crosslinking zone depending on the type of the foaming agent, resulting in inhibition of the crosslinking reaction. Therefore, by extracting the lubricant from the vicinity of the long-land die position at which the degree of crosslinking reaches the desired gel ratio, the smoothness between the long-land die and the resin is decreased and the resistance to the wall surface of the long-land die is increased, thereby applying the back pressure into the long-land die. Even if the foaming agent in the long-land die is partially volatilized or decomposed by the back pressure, the effect that the agent is melted into the resin is achieved.

If the choking device is provided at the desired part of the long-land die 8, 9, 10, further back pressure can be applied. Therefore, it is useful to control the back pressure by co-using the choking orifice 7. The choking orifice is shown in Fig. 4. If desired, a fresh lubricant can be supplied, for again imparting the smoothness to the resin, from the position on the long-land die at which the crosslinking has proceeded sufficiently and the back pressure in the long-land die 8, 9 reached a back pressure sufficient to ensure that the gas formed by volatilization or decomposition of a part of the foaming agent in the long-land die 8, 9 does not foam the resin.

If the lubricant is extracted and not re-introduced the back pressure continues to increase. Therefore, if on reaching the desired back pressure, another lubricant supply flange 6 is provided to supply lubricant through the supply device 18, 19, the back pressure does not increase excessively. This prevents deterioration of shaping properties of the resin crosslinked with increasing the back pressure, but in some cases, the lubricant may not be supplied again depending on the type of resin used.

The back pressure is generally 10 kg/cm<sup>2</sup> or more, preferably 30 to 200 kg/cm<sup>2</sup>. Further, the length of the long-land die 9 necessary to reach the desired back pressure is determined by the shape of resin, the amount of lubricant extracted and the speed of molding, and is generally 30 to 500 cm.

When a chemical foaming agent is used, the crosslinked resin is heated further to a temperature higher than its decomposition temperature at rear part 10, 11 of long-land die.

Length of the rear part 10, 11 of the long-land die 3 necessary to provide decomposition of the foaming agent by heating is preferably 1/5-2 times the length of the front part 6.

The temperature of the long-land die, especially rear part 10, 11, is preferably gradually increased toward the end portion of the die to ensure a uniform, fine cellular structure in both the surface and inner portions of the foamed article.

When physically volatile foaming agents such as butane, pentane, methyl chloride or methylfluoride are used, the agent is added to the resin through inlet 23 provided on the side of extruder 2 as shown in Fig. 1. After kneading and mixing with the resin to form a homogeneous mixture, the

mixture is extruded into long-land die 3.

As shown in Fig. 1, rear part 10, 11 of long-land 3 is preferably configured in a funnel form in which the cross section of the die is gradually enlarged so as to maintain slight pressure on the expanding resin. Most preferably, when the expansion ratio of the resin is more than ten, a die configuration which is arranged to absorb the voluminous expansion stepwise as shown in Fig. 1 is recommended.

According to the present invention, a uniformly expanded, fine-cell foamed article having a low or high expansion ratio, even in a thick article, can be obtained in a continuous process. Notwithstanding the use of a long-land die, the extruding rate of the foamed article is increased over the methods of the prior art, contrary to expectations. Even when the method of the present invention is applied to crystalline polyolefins, which have been deemed to be impossible to form into uniform, fine foamed articles, particularly high density polyethylene and polypropylene, finely foamed articles of a high expansion rate can be obtained.

The following Examples are presented to further illustrate the method of the present invention.

#### Example 1

A masterbatch prepared by mixing and pelletizing 150 parts by weight of a high density polyethylene (Mitsubishi Petrochemical Co., JX20) and 15 parts by weight of a foaming agent (Eiwa Kasei Co., AC #3) and 0.8 part by weight of a crosslinking agent (Nippon Oil & Fats Co., Perbutyl C) were mixed and the resulting mixture was fed into a hopper 1 of an extruder 2 as shown in Fig. 1. The extruder 2 had a diameter of 65 mm and a single screw (L/D = 22). Resin was extruded into front part of a longland die 8 having a cross-section of 20 mm x 200 mm. A thermometer 21 for determining resin temperature mounted at the outlet of extruder 2 indicated 158°C.

Lubricant (Nippon Oil & Fats Co., Nissan Uniblue 75D-3800Z) was supplied constantly from lubricant supplying flange 4 via slit 14 from the side wall at the inlet of front part 8 of the long-land die. Resin was heated at the front part 8 of the long-land die with the temperature distribution of four steps of 170°C, 170°C, 173°C and 175°C along the direction of resin flow. The length of the front part 8 of the long-land die was 3 m. Lubricant extracting flange 5 was provided at the outlet of the front part 8 of the long-land die to remove the lubricant present between the resin in the front part 8 of the long-land die and the die wall surface. The lubricant extracted was discharged to a receiver 17 via slit 15 provided on the inner face of flange 5 and drain pipe 16.

The intermediate part 9 of the long-land die had a length of 1 m and the same cross-section of 20 mm x 200 mm as in the front part of the long-land die. Lubricant supplying flange 6 was provided at the inlet of the intermediate part of the long-land die to again supply the lubricant to the interface between the resin and the wall surface of die.

Resin was heated at the intermediate part 9 of

the long-land die with the temperature distribution of two steps of 176°C and 178°C along the direction of resin flow.

A pressure gauge 17 mounted at the outlet of extruder 2 indicated 43 kg/cm<sup>2</sup>. Further, the gel ratio at the outlet of the intermediate part of the long-land die was 41%.

Resin was heated at the rear part 10, 11 of the long-land die with the temperature distribution of four steps of 180°C, 185°C, 190°C and 190°C to heat decompose the foaming agent. The length of the die was 3 m. Further, the cross-sections of the rear part 10, 11 of the long-land die were 30 mm x 300 mm and 50 mm x 500 mm, respectively. Thus, the rear part is expanded as the resin foams and expands.

Thus, foamed article 12 can be extrusion molded continuously by extruding the resin from the long-land die 11 under atmospheric pressure.

The foamed article according to this example became a continuous foamed board having a rectangular cross-section of 63 mm x 620 mm and was a plate-like foamed article having fine cells having a cell size of 150 microns or less and a skin layer having a density of 0.028 g/cm<sup>3</sup>.

This crosslinked foamed article was heated in a gear oven at 120°C for 15 hours. No dimensional change was observed except slight dolling in the cut edge.

#### Example 2

The same procedure as in Example 1 was followed except that the amount of the lubricant extracted from the lubricant extractor 5 was reduced to half and the lubricant supply from the lubricant supplying flange 6 was stopped. As a result, the same foamed board as in Example 1 could be obtained.

#### Example 3

Using pelletized resins composed of 100 parts by weight of a polypropylene (Mitsubishi Petrochemical Co., FX4) and 18 parts by weight of a foaming agent (Eiwa Kasei Co., AC #3), together with 0.4 part by weight of a crosslinking agent (dibutyl benzene), molding was conducted in the same manner as Example 1 to obtain a plate-like foamed article having a cross-section of 65 mm x 640 mm, fine cells having a cell size of 300  $\mu$  or less and a density of 0.027 g/cm<sup>3</sup>.

#### Example 4

100 parts by weight of a low density polyethylene (Mitsubishi Petrochemical Co., Yukalon LK50), 3 parts by weight of talc and 0.6 part by weight of a crosslinking agent (Nippon Oil & Fats Co., DCP) were mixed and fed into an extruder (L/D = 28, a diameter: 50 mm) wherein a foaming agent inlet 23 was provided on the side of the extruder at a point of 3/5 of the length of the extruder from the front edge of the cylinder so as to introduce a volatile foaming agent (butane) using a gear pump. Resin thermometer 21 mounted at the outlet of extruder 2 indicated 153°C, the overall length of the long-land die was 6 m (front part: 4 m, intermedi-



ate part: 1 m, rear part: 1 m), the temperatures at each part were the same as in Example 1. Further, resin pressure gauge 22 indicated 65 kg/cm<sup>2</sup>.

The front part 8 and intermediate part of the long-land die were a cylindrical die having an inner diameter of 33 mm, and the rear part was a cylindrical die having an inner diameter of 45 mm. Lubricant applicator was provided at the inlet of the front part 8 of long-land die and lubricant was supplied into the interface between the die and resin. Lubricant extracting flange 5 was provided at the inlet of the intermediate part of long-land die to extract the lubricant. Further, lubricant supplying flange 6 was again provided at the outlet of long-land die 10 to supply the lubricant into the interface between the inner surface of die and the resin.

The foamed article 20 extruded from the rear part 10 of the long-land die was a rod-like foamed article having an outer diameter of 86 mm, a density of 0.032 g/cm<sup>3</sup>, gel ratio of 53% and pores having an average cell size of 500 microns.

#### Example 5

100 parts by weight of the high density polyethylene, 15 parts by weight of the foaming agent and 0.8 part by weight of the crosslinking agent which were the same as used in Example 1 were mixed and the resulting mixture was fed into a hopper 1 of extrusion molding machine 2 as shown in Fig. 5. The extrusion molding machine 2 had a diameter of 65 mm and a single screw (L/D = 22). Resin was extruded into the front part 8 having a cross-section of 20 mm x 200 mm of long-land die. Resin thermometer 21 mounted at the outlet of extrusion molding machine 2 indicated 158°C. From the lubricant supplying flange 4, lubricant was supplied constantly via a slit 14 from the side wall of the inlet of front part 8 of long-land die.

Resin was heated at the front part 8 of long-land die with the temperature distribution of four steps of 170°C, 170°C, 173°C and 175°C along the direction of resin flow. The length of the front part 8 of long-land die was 3 m. Choking orifice 7 was provided at the outlet of the front part 8 of long-land die. The structure of choking orifice is shown in Fig. 4 and the cross-section is choked to 2/3. After the choking orifice, the lubricant extracting flange 5 was provided to remove the lubricant present between the resin in the front part 8 of long-land die and the wall surface of the die.

The length of the intermediate part 9 of longland die was 1 m and the cross-section thereof was the same as in the front part 8 of long-land die. Lubricant supplying flange 6 was provided at the outlet of the intermediate part 9 of long-land die to again supply the lubricant to the interface between the resin and the wall surface of the die. The die structure and the temperature conditions of the intermediate part and the subsequent part were the same as in Example 1.

The resin thermometer mounted at the outlet of the extrusion molding machine 2 indicated 50 kg/cm<sup>2</sup>. The foamed article 20 according to this Example was a plate-like foamed article having the properties and cell structure being substantially the

same as in Example 1.

#### CLAIMS

1. A method of producing a crosslinked foamed article comprising uniformly mixing a crosslinkable thermoplastic resin, a foaming agent and a crosslinking agent which has a decomposition temperature higher than the melting point of said resin, extruding said mixture into a long-land die, supplying lubricant into an interface between an inner surface of the long-land die and the resin, decomposing the crosslinking agent by heating to cause crosslinking of the resin, extracting lubricant from the long-land die by a lubricant extracting device at an intermediate and/or an end portion of the long-land die, simultaneously or subsequently heating the foaming agent to the decomposition temperature or boiling temperature thereof, and releasing the resin from the long-land die into an atmospheric or controlled pressure zone to allow the resin to expand and form said foamed article.

2. A method according to claim 1, wherein said lubricant extracting device is provided at an end portion of said long-land die.

3. A method according to claim 1, wherein said lubricant extracting device is provided at an intermediate portion of said long-land die.

4. A method according to claim 3, wherein lubricant is supplied again at a later point in the direction of resin flow after extracting lubricant.

5. A method according to any preceding claim, wherein said thermoplastic resin is a crystalline resin.

6. A method according to any preceding claim, wherein said thermoplastic resin is a polyolefin.

7. A method according to any preceding claim, wherein said foaming agent is a chemical agent which generates gas by decomposition.

8. A method according to any one of claims 1 to 6, wherein said foaming agent is a physical agent which volatilizes by heating.

9. A method according to any preceding claim, wherein a crosslinking agent is decomposed in a front part and a foaming agent is decomposed or heated to its boiling temperature in a rear part of said long-land die.

10. A method according to claim 9, wherein a chemical foaming agent is decomposed in a rear part of said long-land die.

11. A method according to any preceding claim, wherein a rear part of said long-land die has a gradually increasing cross-sectional area along the direction of resin flow.

12. A method according to any preceding claim, wherein a choking orifice is additionally provided in the long-land die.

13. A method as claimed in claim 1 and substantially as herein described.

14. A method of producing a crosslinked foam article substantially as herein described with reference to Figs. 1 to 4 or to Fig. 5 of the accompanying drawings.

15. A method of producing a crosslinked foam article substantially as herein described with refer-

ence to any one of Examples 1 to 5.

16. Apparatus for producing crosslinked foam articles substantially as herein described with reference to Figs. 1 to 4 or to Fig. 5 of the accompanying drawings.

17. A crosslinked foam article when produced by a method as claimed in any one of claims 1 to 15 or in an apparatus as claimed in claim 16.

18. The features as herein disclosed, or their equivalents, in any patentably novel selection.

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